**NUCLEAR DATA AND MEASUREMENTS SERIES**

**ANL/NDM-33**

# Comments on the Energy-Averaged Total Neutron Cross Sections of Structural Materials

by

A.B. Smith and J.F. Whalen

June 1977

**ARGONNE NATIONAL LABORATORY,**
**ARGONNE, ILLINOIS 60439, U.S.A.**

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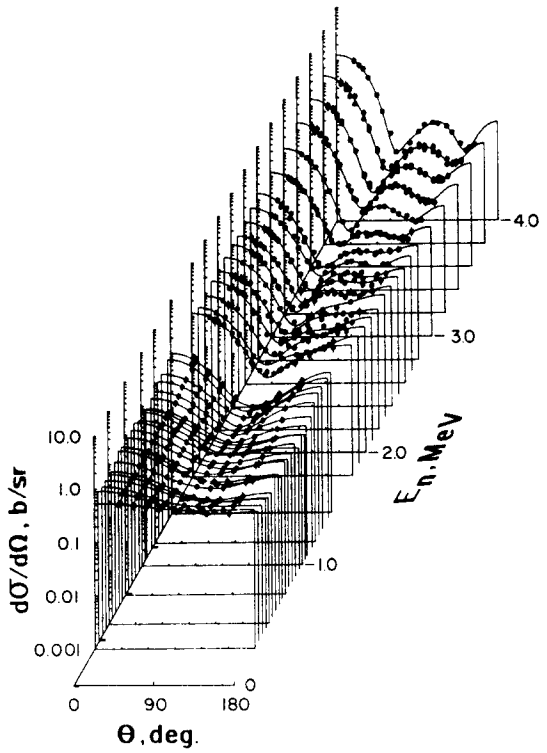


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## NUCLEAR DATA AND MEASUREMENTS SERIES

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# COMMENTS ON THE ENERGY-AVERAGED TOTAL NEUTRON CROSS SECTIONS OF STRUCTURAL MATERIALS IN THE FEW MEV RANGE*

by

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## ABSTRACT

Broad-resolution total neutron cross sections of carbon, iron and titanium are measured with varying sample thicknesses from $\tilde{\sim}$ 1 to 4 MeV. Energy-averaged cross sections are deduced to accuracies of $\lesssim$ 1 percent. The carbon values are consistent with previously reported precision measurements. The measured energy-averaged iron and titanium values are larger than the calculated energy average of some previously reported higher resolution measured and evaluated cross sections by as much as 10 percent. These differences may be indicative of a physical phenomenon which has implications on the design of fast-reactor systems.

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*This work supported by the U.S. Energy Research and Development Administration.

# I. INTRODUCTION

There has been some indication that energy-averaged total neutron cross sections of fluctuating structural materials such as iron and titanium are not well known in the few-MeV range. Integral measurements, such as "broomstick" studies, indicate lower average cross section values (1) than obtained from microscopic measurements. There have been persistent problems with energy-averaged model interpretations (e.g. optical model and statistical model) with discrepancies between models describing differential elastic neutron scattering distributions and energy-dependent total neutron cross sections. The latter discrepancies have become more troublesome as both models and data have become more quantitatively accurate and collective vibrational modes have become of interest. These model discrepancies are difficult to explain in terms of random resonance fluctuations. Some of the total neutron cross section measurements have given primary attention to the resolution of resonance structure. The experimental resolutions employed, while good, may be short of the true resonance detail indicated by theoretical estimates (2). Some of the measurements have given little attention to sample thickness effects and have employed samples that, at the partially resolved resonance peaks, may be several mean-free-paths thick. It has been observed that some of these thicker-sample measurements lead to cross section averages that are systematically lower than other information.

In view of the above questions a "pseudo-integral" measurement was undertaken with the objective of determining the total neutron cross sections of carbon, titanium and iron in the few-MeV range with energy resolutions of 100 to 200 keV. The results are outlined herein and compared

with averages of both evaluated data files and high-resolution measured values. Apparent energy-dependent discrepancies are noted.

## II. SAMPLES

The samples were fabricated into 2.54 cm diameter cylinders of the natural elements with lengths selected to provide a range of average neutron transmissions extending from $\tilde{\sim}$ 20 to 90 percent. Uncertainty in sample dimensionality was limited to within $\leq \pm 0.005$ mm. The carbon samples were machined from pile-grade graphite stock. The iron samples were fabricated from ARMCO stock of >99 percent purity. The titanium stock was of similar purity. Generally, small (<<1 percent) impurities in either iron or titanium samples would have negligible effect upon the present energy-averaged measurements. Atomic-densities were calculated from precision dimensional and weight determinations and the iron and titanium values were consistent with handbook values. The carbon densities were within the range given for pile graphite. It was assumed that the samples were of uniform density. Particularly for titanium, this assumption was supported by the densities of different samples fabricated from various sources of stock material. The sample parameters are presented numerically in Table 1.

## III. MEASUREMENT METHOD

The measurement technique was, in principle, the conventional monoenergetic-source-transmission method modified to provide essentially concurrent measurement of several samples (and voids) as well as background suppression via fast-neutron time-of-flight techniques (3). The $^{7}Li(p,n)^{7}Be$ reaction was used as a neutron source, pulsed for durations of $\tilde{\sim}$ 1 nsec at a repetition rate of 2 MHz. The neutron-energy scale was

controlled by the proton-beam-energy definition to within $\approx$ 5 keV. The incident-energy spread at a zero-degree reaction angle was in the range 125 to 200 keV referenced to the reaction threshold energy. The source was surrounded by a large shield through which a 1 m long, 1 cm diameter cylindrica collimator was inserted at a zero-degree reaction angle. All the samples were fixed to a wheel placed $\approx$ 10 cm from the collimator exit with the sample axes parallel to the collimated neutron beam. The wheel was rotated with a step motion at $\approx$ 50 rpm, sequentially placing samples and voids on the beam line. A 10 cm diameter scintillation detector was placed $\approx$ 5 m from the sample holder on the neutron-beam axis. Pulse-shape-sensitive circuitry suppressed the gamma-ray response of the detector and conventional timing circuitry was used for neutron velocity determination with a resolution of $\lesssim$ 3 nsec. Neutrons passing through the samples were detected as a function of time after burst and correlated with sample position using a digital compute for the control of both sample position and data acquisition. Measurements were made to $\lesssim$ 1 percent statistical accuracy at incident energy intervals smaller than the incident neutron energy spread. The data acquisition system was inclusive of a real-time clock which made possible exact corrections for small dead-time effects. A Monte-Carlo method was used to estimate inscattering perturbations. The corresponding corrections were negligible due to the collimation and geometric configuration. The data was reduced to total cross sections off-line with particular attention to background control. The neutron-velocity resolutions were sufficient to clearly define the time-uncorrelated background and the time-uncorrelated gamma-ray component, and to resolve the primary neutron component from the secondary group resulting from the $^{7}Li(p,n)^{7}Be^{*}$ reaction. The data

reduction procedure was identical for all samples at a given energy excepting the sample-density constants.

## IV. RESULTS AND COMMENTS

### A. Carbon

Over extended energy intervals between 1 and 4 MeV the total cross section of carbon is a relatively slowly varying function of the energy. Thus, measured cross-section values are relatively insensitive to experimental resolution and can be used to test the validity of measurement regimes using various resolutions. This was done in the present measurements using carbon samples with thicknesses providing neutron transmissions extending over the entire range of interest. Sets of carbon measurements were made concurrently with both the iron and titanium measurements and the data were reduced to cross sections in an explicitly identical manner (e.g. identical resolutions, backgrounds, dead-time corrections, etc.). The results obtained with the three sample thicknesses of 1, 3, and 5 cm were identical within statistical accuracies in the regions of smoothly varying cross sections. Illustrative results are compared with the results of Schwartz et al. in Fig. 1 (4). The energy scales are consistent and comparison of results near the 2.08 MeV resonance supports the estimates of the resolution width of the present experiments. Results obtained with the 3- and 5-cm samples were generally consistent to within <1 percent; the largest differences tended to be in the regions of resonances, as expected. In the smoothly varying regions the present results are consistent with those of Schwartz et al. to within $\lesssim$ 1 percent. A possible exception is the region 2.3 to 2.5 MeV where the present results are 1-2 percent lower than those of Ref. 4; a similar small trend has

been noticed on several past occasions. Results, such as illustrated in Fig. 1, give confidence to the present measurements. Values obtained with widely different transmissions are consistent with one another and with precision results obtained elsewhere using very different resolutions and techniques. The wide variation in sample sizes of the present experiments did not expose such possible experimental errors as improper dead-time correction, background estimate, etc. Thus the carbon results reasonably support the specified accuracies of the present measurement techniques.

B. Titanium

The titanium measurements were made with a 125- to 135-keV incident-energy resolution (referenced to the $^{7}Li(p,n)$ threshold energy) and included the three titanium and three carbon samples of Table 1. Measurements were made at average incident-neutron-energy intervals of $\tilde{\sim}$ 50 keV from 1.0 to >4.0 MeV. The measured transmissions were reduced to cross sections with the results shown in Fig. 2. The cross section values fluctuate with energy. The thinnest sample (2 cm) resulted in very large transmissions with commensurately larger statistical-cross-section uncertainties. The uncertainties for the thicker 3 cm sample were smaller, and those for the 6 cm sample <1 percent. The differences between results obtained with the three sample thicknesses vary with energy as the underlying resonance structure changes. However, there is a systematic tendency for the thinner-sample results to be the larger. This trend decreases with increasing energy to negligible differences at energies above 4.0 MeV. Clearly, these broad resolution cross section values are sensitive to sample thickness. The results were corrected to zero-sample thickness by linearly fitting the values for various sample

thicknesses at each energy and extrapolating to zero thickness. The resulting zero thickness cross sections are shown in Fig. 3. The correction to zero-thickness values varied with energy but when averaged over $\tilde{\sim}$ 250 keV the trend of the corrections ranges from a -0.5 to -1.0 percent change in cross section per cm increase of sample thickness at 1.0 to 2.0 MeV to very small values above 4.0 MeV. Presumably, at the higher energies the true resonance structure has considerably broadened and the cross section is tending toward a relatively smooth behavior with the consequence of reduced sensitivity to sample thickness. This smoothing is enhanced by the multi-isotopic nature of the natural element. This extrapolation procedure is reasonably valid in the range of these samples as discussed below in the context of iron. Exact experimental comparisons should employ average transmissions obtained for identical sample thicknesses. High resolution data reported in the literature generally involved sample thicknesses much greater than used in the present broad resolution measurements making such direct comparison difficult.

The above broad-resolution results can be compared with the reasonably-documented and much higher-resolution titanium results of Schwartz et al. (4). In doing so, the latter values must be averaged and there are uncertainties in that procedure due to a lack of detailed knowledge of the respective resolution functions. For the present purpose simple square resolution functions with widths of 100 and 150 keV are used, centered about the average energies of the present measurements. These two widths generally cover the range of resolutions employed in the present experiments. The two averages of the results of Ref. 4 are similar as shown in Fig. 3. However, at energies below 3.0 MeV both of these averages are lower than the cross

sections of the present broad resolution measurements by amounts of 5 to 10 percent. The discrepancy decreases with increasing energy and for energies of $\gtrsim$ 4.0 MeV it is a marginal 2 to 4 percent. The differences can not be attributed to errors in sample density, dead-time correction, etc., in the present measurements, since such distortions would tend to be relatively constant with energy. The measurements of Ref. 4 employed a 10.7 cm thick sample. This is a very thick sample. Using the cross-section results of Ref. 4, the transmissions of a 6 cm sample were calculated, averaged and compared with the present broad resolution results as illustrated in Fig. 4. The discrepancy between the present measured transmissions and those constructed from Ref. 4 is $\approx$ 10 percent. Similar comparisons using 2 and 3 cm sample thicknesses showed even larger discrepancies. Thus, while the results of Ref. 4 may be very precise in the context of the thick-sample employed and the given experimental resolutions, the deduced cross sections appear to differ from the true values by large amounts and are not suitable for reproducing broad-resolution results obtained with other (and thinner) samples. The sample-size effects are mitigated by improved experimental resolution and theoretical estimates indicate the titanium resonance extrema in the low-MeV-energy region are much larger than experimentally resolved in Ref. 4 (2,5). Quantitative measurement of the cross section in this context requires attention to sample thickness and/or experimental resolution. A similar problem may exist in many of the measurements of total-neutron cross sections of other similar nuclei; e.g. iron, nickel, etc. The magnitude of the effect may well depend upon the details of the particular experimental procedure and the true resonance structure. Neither are

particularly well known. Many of the reported measurements are too poorly documented to make possible an assay of the possible scope of the problem. Quantitative theoretical estimates of true resonance structure and its impact on experimental measurements are seldom made. Preliminary studies, to be reported elsewhere, suggest that the effects can be large (2). This titanium example suggests that total neutron cross sections of a number of structural materials of applied interest may be systematically distorted toward too low values to the detriment of applied calculations and to the confusion of theoretical interpretations.

C. Iron

The above titanium measurements were extended to iron. The incident neutron resolution was a broader $\tilde{\sim}$ 200 keV and the measurements were truncated to the energy range 1.0 to 3.0 MeV. A wider range of sample thicknesses was employed as defined in Table 1. Again, carbon cross sections were concurrently determined in order to test the fidelity of the apparatus. The results are outlined in Fig. 5. The sample thickness effect is very clearly evident with the thin (2 cm) sample results being approximately 10 percent larger than those obtained with the thickest sample (8 cm). The difference decreases with increasing energy. The zero-thickness cross section values were obtained by linear extrapolation as for the titanium case, above. Again, this procedure was recognized as an approximation reasonably valid for only these samples. The correction factors were in the order of 1 percent per cm of sample thickness decreasing with increasing energy. The corrected results are outlined in Fig. 6. The correction to broad resolution results using relative thin samples ($\tilde{\sim}$ 1 cm) is not negligible and there are references in the literature to very much thicker samples (e.g. >10 cm).

The above broad resolution results were compared with average values of the total cross section of iron as given in the ENDF/B-IV (MAT-1192) file (6). Again the exact details of the resolution functions were uncertain therefore a square-resolution average of the file was used based upon the 200 keV resolution of the present experiments at the $^{7}Li(p,n)^{7}Be$ threshold. Subsequent conclusions were not sensitive to the choice of the averaging increment with averaging widths of 100, 200 and 300 keV leading to the same qualitative conclusions. At incident energies of $\lesssim$ 1.5 MeV the magnitudes of the present cross section results and the averages constructed from ENDF/B are similar as illustrated in Fig. 6. The discrepancies are generally a few percent and approximately equally distributed between negative and positive effects. Qualitatively, the differences can easily be attributed to uncertainties in the averaging procedures. Above $\gtrsim$ 1.5 MeV the differences between the present results and the ENDF/B averages become quite systematic with the present results being 4 to 6 percent larger. In no case do the present results fall below the ENDF/B average and in some regions they are 8-9 percent higher. The discrepancy appears somewhat energy dependent being smaller at higher energies. These discrepancies are also present in comparisons of measured transmissions of a 4 cm sample with those calculated from an average of transmissions derived from the ENDF/B file as illustrated in Fig. 7. Similar transmission comparisons at other sample thicknesses (e.g. 2 and 8 cm) show similar discrepancies that tend to be sample-thickness dependent.

The ENDF/B file is an evaluation involving judgements as to the validity of various microscopic data sources. Thus traceability to specific measurements, sample thicknesses, resolutions, etc., is diffi-

cult. However, several general considerations appear applicable. The best resolution measurements available for this file generally involved velocity-measurement techniques. The energy resolution of such measurements deteriorates with increasing energy. Iron is essentially monoisotopic and in the few-MeV region the resonance structure is known to be pronounced. Theoretical estimates may lead to resonance extrema much larger than observed experimentally (2). At least some high-resolution iron measurements have employed very thick samples; in one case 11.8 cm. These considerations and the present measurements tend to suggest that there were some significant distortions in the commonly used iron total cross section analogous to those appearing in titanium, above. The fact that they are not evident at low energy (1.0 to 2.0 MeV) may reflect the great attention that has been given to the resolution in experimental measurements at these lower energies. At increased energies the resolution is less satisfactory and the true structure more complex. Thus the situation may have tended toward the titanium results discussed above.

Very recently Harvey et al. (7) have measured iron total neutron cross sections with improved experimental resolutions. Their sample was relatively thick but the experimental resolution was several keV or better in the energy region of present interest. These new high resolution results were averaged in the same manner as outlined above in the context of ENDF/B and compared with the present broad-resolution values. The present 4 cm thick-sample results are most directly comparable with the Harvey et al. values obtained using a 5.08 cm sample. The present transmissions observed with the 4 cm sample are compared with an equivalent average constructed from the Harvey et al. results in Fig. 7. Above $\approx$ 1.5 MeV

the two sets of measured values agree, on the average, to much better than 1 percent. At lower energies the differences are randomly a little larger probably reflecting some detailed differences between the averages used in comparing the two sets of measurements. Similar good agreement was obtained for transmissions at other sample thicknesses (e.g. 2 cm and 8 cm). Furthermore, the present zero-thickness cross sections are in very good agreement with the equivalent average values constructed from the data of Harvey et al. as shown in Fig. 6. Above 1.5 MeV the average cross sections obtained from the two measurements generally differ by less than 1 percent. These various comparisons indicate that results obtained with sufficiently good resolutions to resolve the majority of inherent resonance structure, as in the work of Harvey et al., are consistent with precise broad resolution results both in cross section and in transmissions as observed over a wide range of sample thicknesses. Both the present results and the energy averages of the Harvey et al. results suggest that ENDF/B-IV energy-averaged total-cross-section magnitudes are generally too low by 4-6 percent over the energy range 1.5-3.0 MeV. Over this energy range none of the energy-averaged results derived from either set of measurements are significantly less in magnitude than comparable average values constructed from ENDF/B-IV and in some broad energy regions (e.g. about 2.3 MeV) are 8-10 percent higher than ENDF/B-IV.

## V. CONCLUDING REMARKS

In the few-MeV range the total cross section of iron is approximately 60 percent elastic scattering. Thus a 5 percent increase in the total cross section could enhance the elastic scattering by $\tilde{\sim}$ 8 percent assuming

the non-elastic cross section remains constant and internal consistency of the file is maintained, as usually required. FBR cores contain appreciable amounts of iron arranged in relatively thin elements and their sensitivity to the elastic scattering cross sections of iron is significant. For example, the sensitivity of $k_{eff}$ of the ZPR3-48 fast critical assembly to changes in the iron elastic scattering cross section in the 1.0 to 3.0 MeV range is about $0.6 \times 10^{-2}$ percent change in $k_{eff}$ per percent change in cross section (8). Thus, if the above iron measurements are correct the consequences could be an increase in reactivity of as much as $\tilde{\sim}$ $\delta k/k = 0.0005$ relative to that calculated using ENDF/B-IV. ENDF/B-IV calculations characteristically underpredicted k by $\tilde{\sim}$ 0.005 thus the possible changes in the iron cross section could account for $\tilde{\sim}$ 10 percent of the discrepancy between $k_{eff}$ as calculated and measured. Such changes are about half those obtained with reasonable changes in the fission cross sections (9). Large FBR designs have involved iron shields $\gtrsim$ 60 cm thick (1). Five percent changes in the total cross section, as suggested above, will change the direct uncollided neutron penetration of few MeV neutrons through such a shield by factors of 2 to 3. This change is in the opposite direction from that suggested by some integral measurements (1). However, this is not a contradiction as thick-shield penetration is largely influenced by the minima of the cross section and not the average magnitude. Indeed, incomplete resolution of extremia (minima or maxima) is a probable source of the discrepancies in averaged-cross-section magnitudes noted above.

Global nuclear model calculations founded upon higher-energy elastic distributions (e.g. the optical potential derived in Ref. 10) frequently do not portray the average value of the total cross section near the diffraction minima at $\tilde{\sim}$ 1.0 MeV and A = 40-60. The calculated total cross sections tend to be larger than indicated by experiment and it is difficult to mitigate the problem with energy dependent potential parameters. Moreover, one would expect the total cross section to be one of the less ambiguous calculated quantities. If, as suggested above, the measurements in the region A = 40-60 and energy 1.0-3.0 MeV have often resulted in energy-averaged total cross sections that are too low by 5 to 15 percent a large share of the discrepancy between measured and calculated values vanishes.

Finally, the above suggests that the first "benchmark" test of a fine resolution microscopic data file could well be a broad resolution microscopic measurement. Without good agreement between the fine and broad resolution microscopic data it seems difficult to justify the far more complex and ambiguous macroscopic "benchmark" tests.

## ACKNOWLEDGEMENTS

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TABLE 1. Sample Parameters

| Element | Diameter (cm) | Length (cm) | At/Cm$^2$ ($\times 10^{-23}$) | Transmission[a] |
|---|---|---|---|---|
| C-1 | 2.54 | 1.0 | 1.129 | 0.798 |
| C-2 | 2.54 | 3.0 | 3.390 | 0.508 |
| C-3 | 2.54 | 5.0 | 5.645 | 0.323 |
| Fe-1 | 2.54 | 2.0 | 1.696 | 0.601 |
| Fe-2 | 2.54 | 2.5 | 2.120 | 0.529 |
| Fe-3 | 2.54 | 4.0 | 3.392 | 0.362 |
| Fe-4 | 2.54 | 8.0 | 6.784 | 0.131 |
| Ti-1 | 2.54 | 2.0 | 1.132 | 0.712 |
| Ti-2 | 2.54 | 3.0 | 1.698 | 0.601 |
| Ti-3 | 2.54 | 6.0 | 3.396 | 0.361 |

[a]Illustrative transmissions assuming $\sigma_T$ = 2 b(C), = 3 b(Ti and Fe).

## FIGURE CAPTIONS

Fig. 1. Illustrative total neutron cross sections of carbon. The line represents the fine resolution measurements of Schwartz et al. (4). Data points are the present results obtained with $\approx$ 200 keV resolution and 3 cm (circular points) and 5 cm (cross points) sample thicknesses.
(ANL Neg. No. 116-77-7)

Fig. 2. Titanium total neutron cross sections obtained with $\approx$ 125 keV resolution and 2(0), 3(Δ) and 6(+) cm thick samples.
(ANL Neg. No. 116-77-2)

Fig. 3. The present titanium total cross section results obtained with $\approx$ 125 keV resolution corrected to zero sample thickness (0). 100 keV and 150 keV averages of the results of Schwartz et al. (4) are indicated by dashed and solid curves, respectively.
(ANL Neg. No. 116-77-6)

Fig. 4. Neutron transmissions through a 6 cm titanium sample. The present broad resolution results are indicated by data points. 100 (---) and 150 (---) keV averages constructed from the cross section values of Ref. 4 are indicated by the curves.
(ANL Neg. No. 116-77-173)

Fig. 5. Iron total neutron cross sections obtained with $\approx$ 200 keV resolution and 2(0), 2.5(Δ), 4(+), and 8(X) cm thick samples.
(ANL Neg. No. 116-77-3)

Fig. 6. Total neutron cross sections of iron. The present broad resolution measured values are indicated by data points (0). Equivalent average values constructed from the measured values of Harvey et al. (7) are also indicated by data points (X). The respective average of ENDF/B-IV is indicated by the solid curve. (ANL Neg. No. 116-77-171)

Fig. 7. Neutron transmissions through a 4 cm iron sample. The present broad resolution results are noted by (0), equivalent averages constructed from the data of Harvey et al. by (X). The solid curve is the energy-averaged transmission as constructed from ENDF/B-IV.
(ANL Neg. No. 116-77-172)

Fig. 1

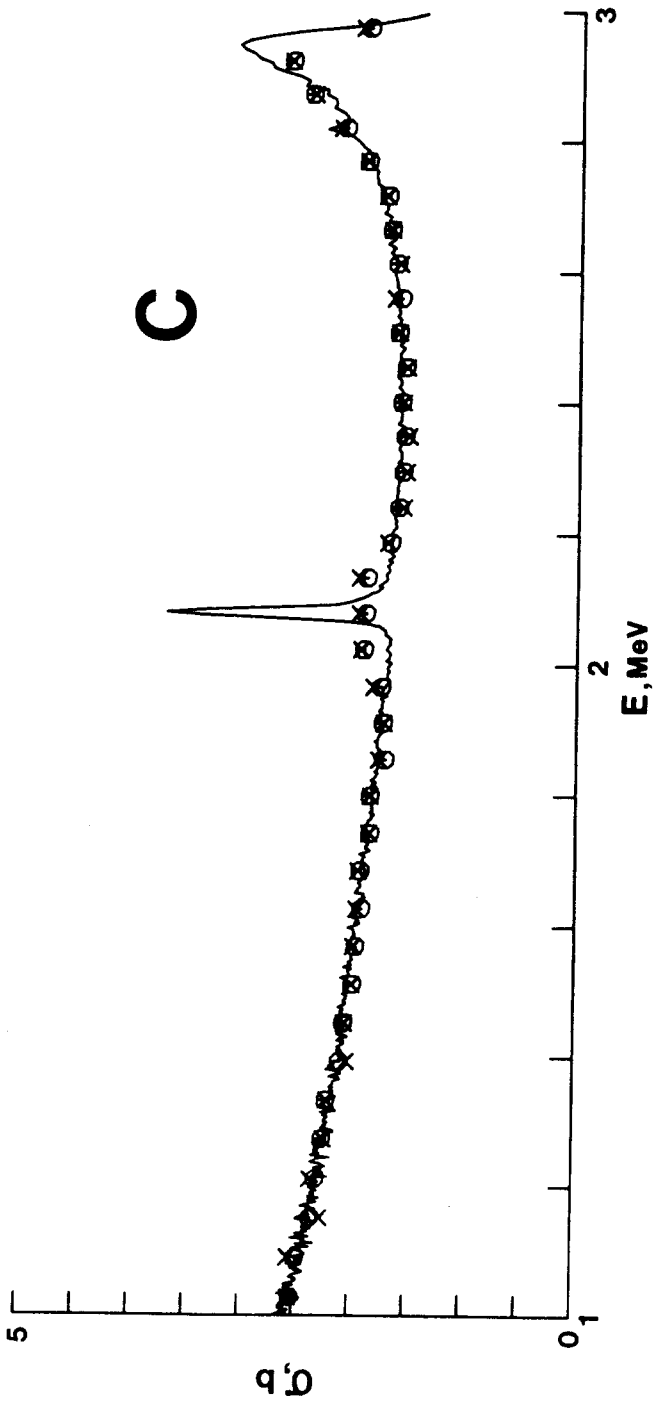

Fig. 2

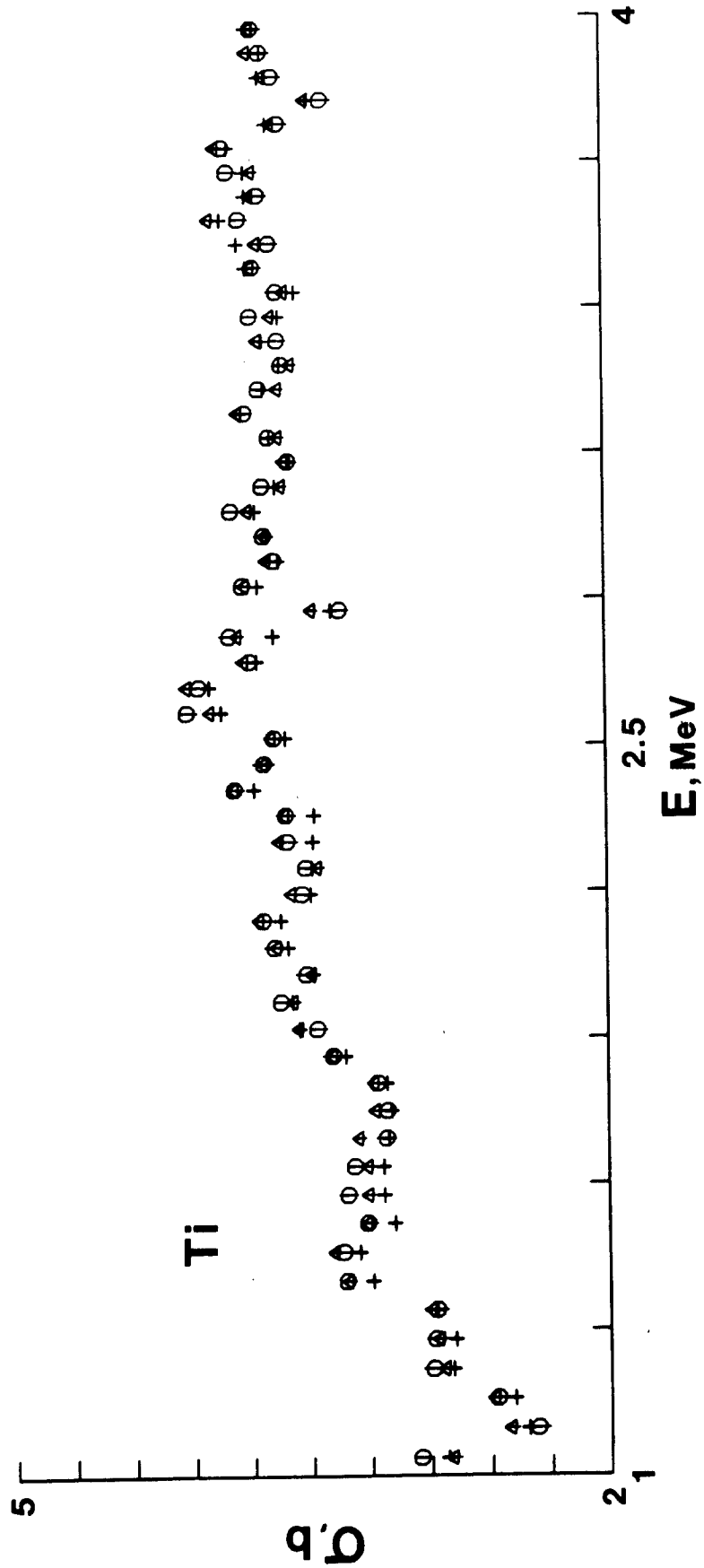

Fig. 3

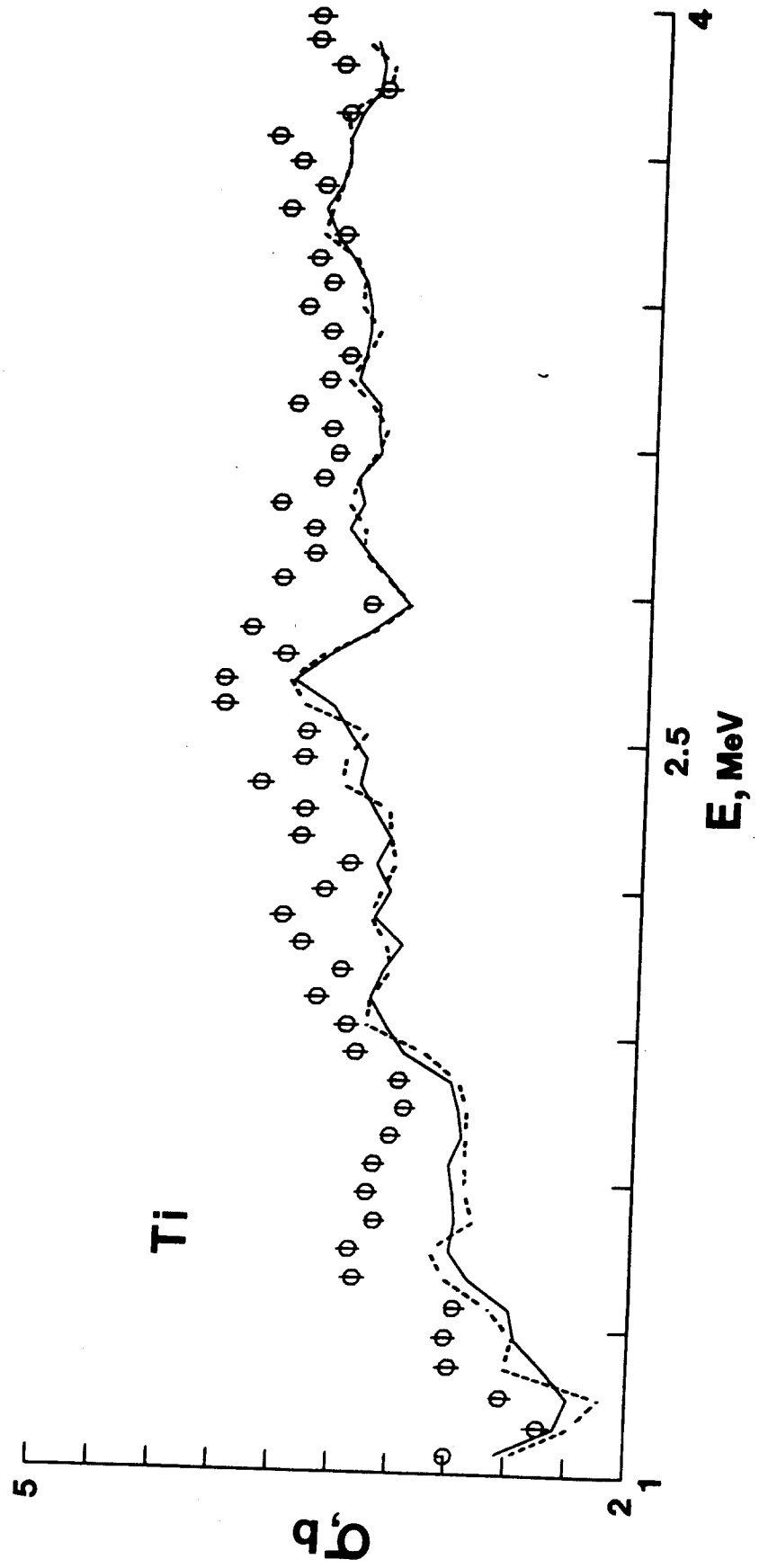

Fig. 4

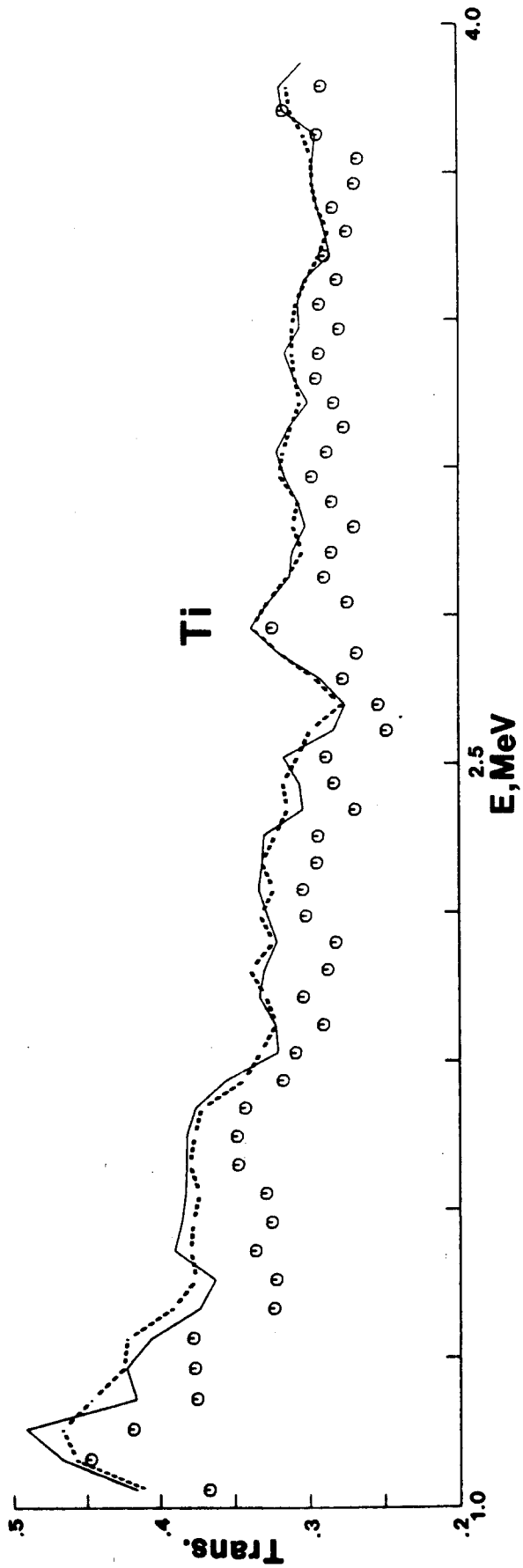

Fig. 5

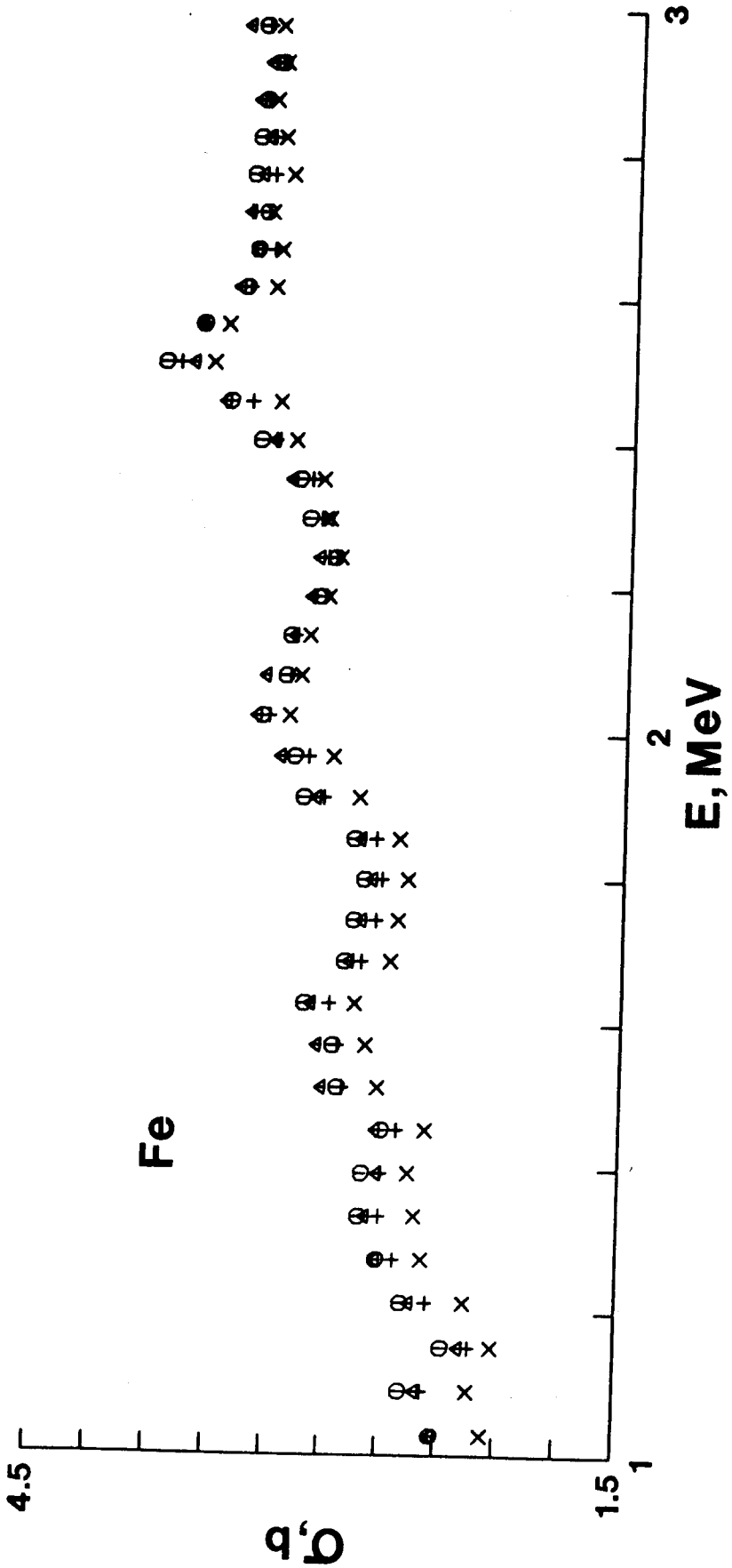

Fig. 6

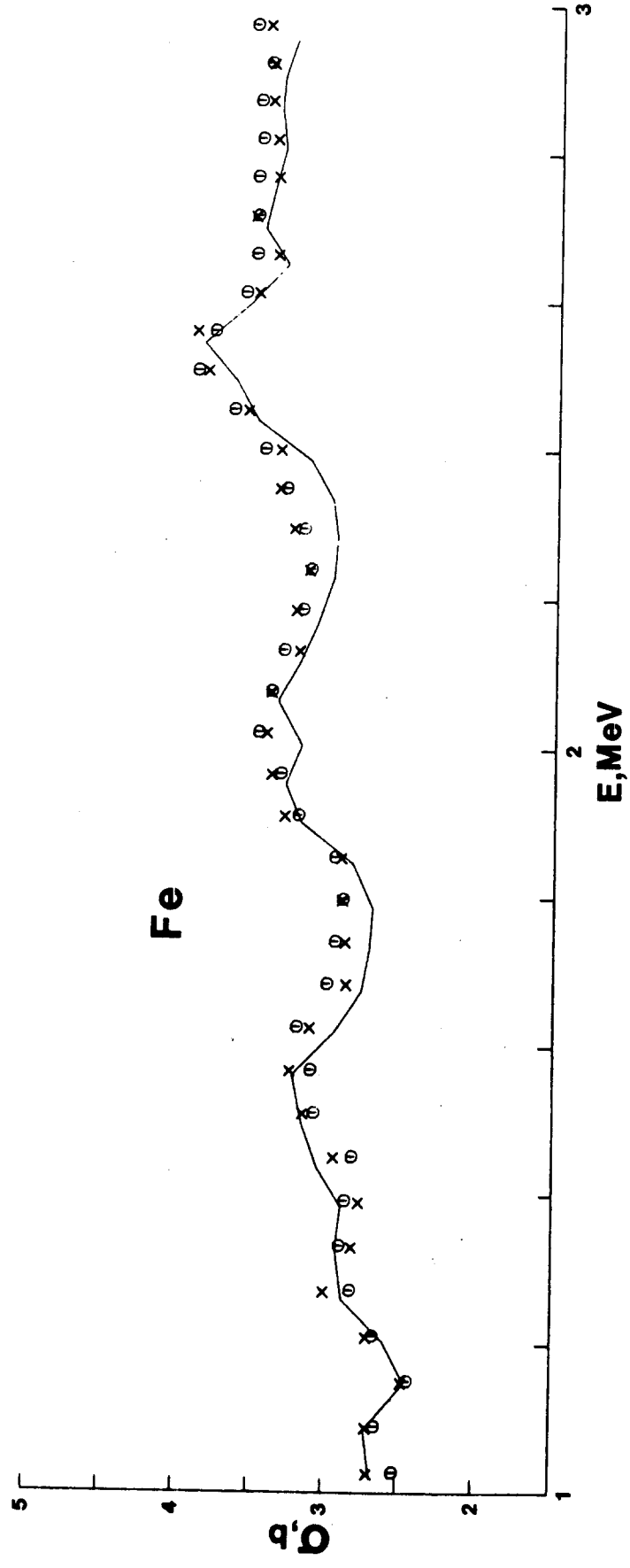

Fig. 7